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Drivers of contaminant levels in surface water of China during 2000–2030: Relative importance for illustrative home and personal care product chemicals

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ABSTRACT

Water pollution are among the most critical problems in China and emerging contaminants in surface water have attracted rising attentions in recent years. There is great interest in China's future environmental quality as the national government has committed to a major action plan to improve surface water quality. This study presents methodologies to rank the importance of socioeconomic and environmental drivers to the chemical concentration in surface water during 2000–2030. A case study is conducted on triclosan, a home and personal care product (HPCP) ingredient. Different economic and discharge flow scenarios are considered. Urbanization and wastewater treatment connection rates in rural and urban areas are collected or projected for 2000–2030 for counties across China. The estimated usage increases from ca. 86 to 340 t. However, emissions decreases from 76 to 52 t during 2000–2030 under a modelled Organisation for Economic Co-operation (OECD) economic scenario because of the urbanization, migration and development of wastewater treatment plants/facilities (WWTPs). The estimated national median concentration of triclosan ranges 1.5–8.2 ng/L during 2000–2030 for different scenarios. It peaks in 2009 under the OECD and three of the Intergovernmental Panel on Climate Change (IPCC), A2, B1 and B2 economic scenarios, but in 2025 under A1 economic scenario. Population distribution and surface water discharge flow rates are ranked as the top two drivers to triclosan levels in surface water over the 30 years. The development of urban WWTPs was the most important driver during 2000–2010 and the development of rural works is projected to be the most important in 2011–2030. Projections suggest discharges of ingredients in HPCPs - controlled by economic growth - should be balanced by the major expenditure programme on wastewater treatment in China.

1. Introduction

Water contamination can be harmful to human and ecosystem health. Emerging contaminants, such as pharmaceuticals, home and personal care products (HPCPs) have raised growing concerns (Boxall et al., 2012; EPA, 2017). Reducing untreated wastewater and protecting aquatic ecosystems are targets of the Sustainable Development Goals set by the United Nations to be reached by 2030 (Hering et al., 2016). China is a country with major challenges of water quality and availability, including: the size and diversity of the country and its rivers; the population size and migration; rapid economic growth, with increased industrial, agricultural and domestic demands for usable water and the effluents that these activities generate. The Chinese Government therefore developed an 'Action Plan for Water Pollution Prevention' in mid-2015 (MEP, 2015a), which is laid out in the national 13th 5-Year Plan (CPGC, 2016). However, in order to make rational, effective and informed decisions which will improve water quality, there is an urgent need for a methodology to identify the potential key drivers to affect the level of contaminants in surface water, especially for those anthropogenic-source contaninants. The results could improve and inform understanding, policy and decision-making.

Socio-economic activities and environmental changes have affected water quality in China during the remarkable development over the past decades (Gleick, 2008–2009). Ingredients in HPCPs represent an

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interesting case, because they are closely linked to socio-economic activities and as trace organics they could be markers of sewage and anthropogenic-source ingredients (Gasser et al., 2010; James et al., 2016). Some HPCPs are relatively poorly studied so far and have attracted increasing interest in recent years (Boxall et al., 2012) such as UV filters and parabens, yet some of them are abundant and being considered for environmental limits and more controlled usage, such as triclosan and triclocarban. HPCPs are diffusively discharged in wastewaters. Their consumption increased between ~40–~800% in China during the economic boom between 2000 and 2012 (Euromonitor, 2015); however, China also increased its wastewater treatment capacity by about 8-fold during the same period (Supporting Information (SI) Fig. S1), due to several factors (i.e. rising urbanization, compliance with discharge standards) (MHURD, 2013), potentially counterbalancing the potential release of HPCPs to the environment. Other marked societal and infrastructure trends have occurred and will continue in future. Population growth and migration and rapid urbanization progress (Yang, 2013) across China make the change of ingredient usage and release more complex geographically. Changing discharge flow, linked to environmental and infrastructure changes, also affects the dilution of chemicals in surface water. These factors have changed/are changing in a way that could be strongly impacting the concentrations and distributions of chemicals in surface water and the water quality (Zhu et al., 2016; Zhu et al., 2014).

This study was therefore conceived to develop a modelling approach, to explore the potential influence of several key drivers on past, present and future chemical surface water concentrations in China. We address several drivers which will influence ingredient usage, release and loading in aquatic systems, namely economic development as Gross Domestic Product (GDP), population, urbanization, wastewater treatment capacity and discharge flow rates, to estimate temporal changes in water concentrations. Measurements of pharmaceutical and HPCP ingredients have only become available for a limited number of regions in recent years in China. We chose triclosan as an example ingredient in the calculation as it is well studied and there are more monitoring data available than other ingredients for model validation (Zhu et al., 2016). It enters the aquatic environment diffusively, primarily from domestic wastewaters. Scenarios considered here take account of main drivers discussed above to: (1) model usage, emissions and concentrations in surface water of triclosan in China as an example ingredient between 2000 and 2030, assuming that it continues to be used in HPCPs in that period; (2) identify and rank the key drivers affecting surface water concentrations. This approach has not been used before, but we believe it can be adapted and applied to a range of chemicals from anthropogenic sources with different usage/release scenarios in future, using the base data and modelling tools assembled here.

2. Methodologies and approaches

2.1. Ingredient usage under five economic scenarios

OECD (Organisation for Economic Co-operation) per capita GDP (OECD, 2014) was found to significantly correlate ($R^2 > 0.88$) to sales volumes (tonnes) of HPCP categories which contain triclosan (SI Table S1 and Fig. S2) (Mintel, 2014) in the Chinese market for 2000–2019 (Euromonitor, 2015). As seen in SI Fig. S2, most correlations fit linear regression, except those for shampoo, bleach/disinfectant and all-purpose surface care products (SI). Besides these fast moving consumer HPCP categories in Table S1, triclosan is also used in plastic materials, textiles, surface of medical devices, etc. These usages and releases were not taken into account in this study as they are expected to be a small proportion of the total (Euromonitor, 2015; SCCS, 2010). In addition, emission pathways are complex depending on how these materials are disposed of; and triclosan leaching from these materials is likely to be slow (SCCS, 2010) compared to the daily used HPCP categories.

Based on above regressions, the annual sales volumes of HPCP categories which contain triclosan were extrapolated for 2011–2030 under five economic scenarios, i.e. one from OECD future GDP outlook (OECD, 2014) and four predicted by the Centre for International Earth Science Information Network (CIESIN) under four IPCC marker scenarios (A1, A2, B1 and B2) (CIESIN, 2002). OECD is an authorivative economic institution that could provide potentially reasonable economic outlook as reference. IPCC scenarios consider extreme and moderate conditions, which have been widely used in ecology or energy relevant studies. For the feature of the five economic scenarios and their comparison see SI and Table S2. The two 'A' economic scenarios were extreme; the two 'B' scenarios and the OECD projection are moderate (SI Fig. S3). The historical sales volume for 2000–2010 was directly taken from the Euromonitor database (Euromonitor, 2015). Products sold in a year were assumed to be consumed within the same year. Triclosan usage was, therefore, estimated from product sales volumes, the inclusion level in products and the percentage of product variants (Mintel 2014) that contain triclosan for all product categories (Eq. (1) in SI). The product categories and triclosan inclusion level (0.3%) (Hodges et al., 2012) were assumed identical during 2000–2030. Based on the usage, emission and surface water concentrations were estimated under these five economic scenarios with the projection of other main drivers described below.

It is assumed there will be no bans or elimination of triclosan in this study till 2030 in China. However, future replacement or reduction of ingredient use is possible due to potential government restrictions or if it is phased out by industry.

2.2. Population and urbanization

The gridded (~1 km) Chinese population count and density were projected by CIESIN for the years 2000, 2005, 2015 and 2020 (CIESIN, 2016a, 2016b) and by another study for 2030 (Feng and Qi, 2016; Qi et al., 2015). For the year 2010, to keep identical to our previous study (Zhu et al., 2016), data projected by Landscan (Landscan, 2010) was used. The urban population was identified by the population density > 5000 (CIESIN, 2011) cap/km^2 for 2030 and > 1000 cap/km^2 for the other 5 years. 5000 cap/km^2 was suggested in some publications but found too high on the population data by CIESIN. Therefore, 1000 cap/km^2 was additionally used in this study, which could result in more reasonable results since they are verified with the census of the national urban population (CNSTATS 2000–2015) (SI Fig. S4) and the predicted potential annual urban population growth rate for future China (ca. 2%). The projected urban expansion from 2000 to 2030 is shown for every five years in Fig. 1. To fill the population data gaps between every two adjacent years with existing projected data (e.g. years between 2000 and 2005), it was assumed that the annual change rate of urban and rural population would be steady between the two years. The projected population was applied to the five economic scenarios to spatially allocate ingredient usage and to estimate population connectivity to wastewater treatment facilities installed in rural and urban areas during 2000–2030.

2.3. Wastewater treatment connection rates

The records for estimating the wastewater treatment connection rates, i.e. the proportion of population connected to wastewater treatment, could be found from the yearbook for 2002–2013 (covered by light green shade areas in Fig. 2A) for urban areas and for 2008–2013 for rural areas for China (MHURD, 2013). The wastewater treatment connection rate was estimated by total wastewater discharge volumes dividing those volumes treated for individual cities. The average of city level wastewater treatment connection rates for urban areas increased from 40% to 89% between 2002 and 2013; the average of provincial level wastewater treatment connection rates for rural areas increased from 2.5% to 6.4% between 2008 and 2013 (Fig. 2A) (MHURD, 2013). Nationally, urban wastewater treatment plants (WWTPs) developed

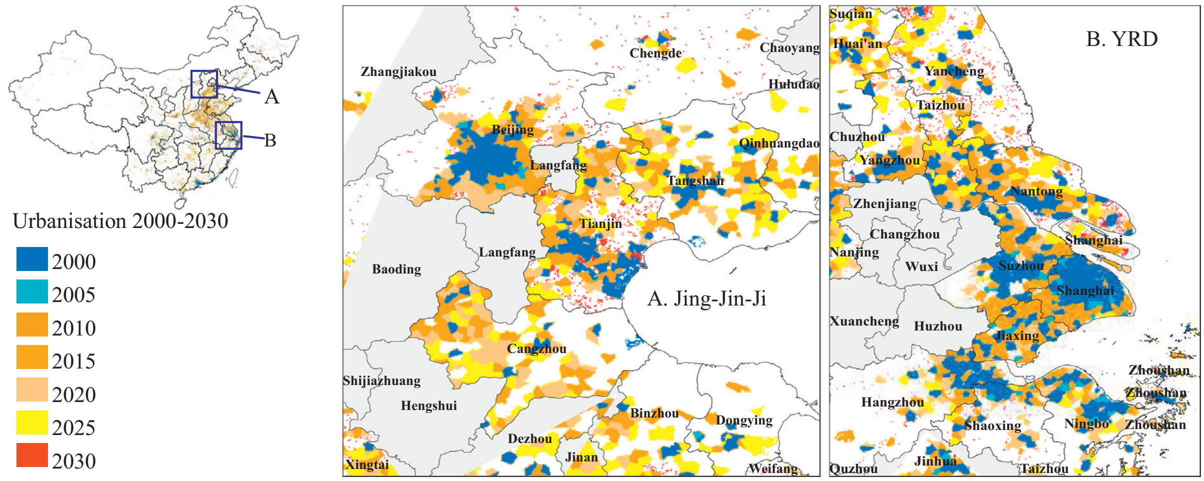

Fig. 1. Illustrative urban expansion information every five years for 2000–2030 in 2 regions; A. Jing-Jin-Ji: Beijing-Tianjin-Hebei; B. Yangtze River Delta (YRD). The different colours represent the urban population (for 2000) and the new urban population compared to that five years ago (for 2005–2030).

most rapidly during 2003–2009 with the total national treatment capacity increasing from ca. 0.3 to 1 billion m^3 water/day (SI Fig. S1) (MEP, 2015b). After 2009, the rate of increase decelerated, but the national investment in WWTP construction was stable during 2009–2013 (MHURD, 2013). Therefore future treatment rates during 2014–2030 were assumed to increase at the same annual rate of change as that during 2009–2013 for urban areas for individual city and the same as that during 2008–2013 for rural areas for individual province. Collected historical and predicted future wastewater treatment connection rates for urban and rural areas were used in all five economic scenarios for estimating the proportions of domestic sewage treated by wastewater treatment infrastructures for each county. More details see SI.

2.4. Releases of ingredient to the environment

As stated above, the use of triclosan is mostly in HPCPs, which is then released with wastewater to the environment directly or via WWTPs with wastewater effluent to the aquatic environment. The possible release to soil by wastewater irrigation and sludge application regionally was not considered in this study due to the lack of information. It is the same assumption made in the previous study by Zhu et al. (Zhu et al., 2016). National triclosan usage was spatially allocated to counties across China by population for each year during 2000–2030 with assumptions that the usage per capita was spatially constant across China for individual years. The usage by respective urban and rural populations was then estimated by urban and rural population for each county. The wastewater generated by respective urban and rural populations was linked to WWTPs proportionally as the treatment rates estimated above.

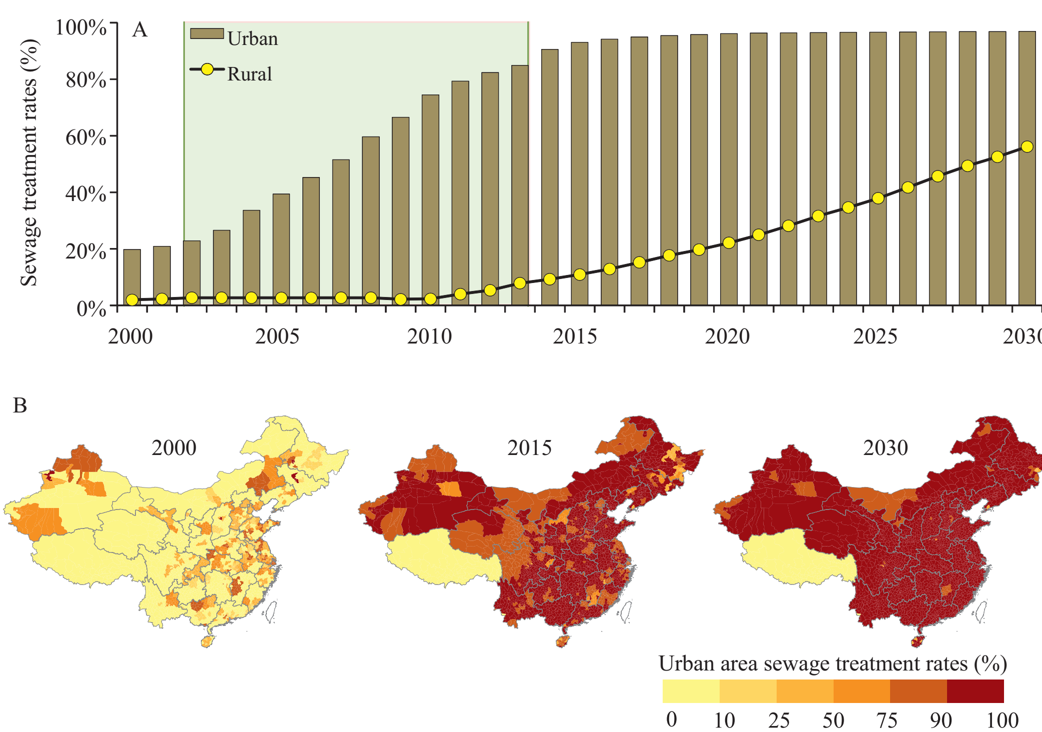

Fig. 2. A. collected (2002–2013, covered by light green shade area) and predicted national average urban and rural wastewater treatment connection rates during 2000–2030; B. the distribution of wastewater treatment connection rates in urban areas at city level across China for 2000, 2015, 2030.

The measured removal efficiency of triclosan in urban WWTPs has been reported to vary between 35 and 98% as a result of different loading mass, treatment technologies, operation conditions, sampling seasons and methods etc. (Agüera et al., 2003; Bendz et al., 2005; Bester, 2003; Heidler and Halden, 2008; Lozano et al., 2013; Ying and Kookana, 2007). In rural areas in China, decentralized sanitation systems (household or neighbourhood scale) or centralized wastewater treatment facilities (village level) are used for different conditions (Godavitarne et al., 2011; Qiang et al., 2013). Many studies show that the removal efficiencies of triclosan by constructed wetlands used in rural areas could range 70–100% (Zhao et al., 2016), but the removal efficiency by other facilities/systems used for rural areas in China has not been reported. Due to a lack of information on spatial distribution of different urban WWTPs and rural facilities as well as low wastewater treatment connection rates in rural areas, a removal efficiency of 95% predicted by SimpleTreat 3.2 (Franco et al., 2013) to represent processes in secondary activated sludge plants (most common technology in China (Jin et al., 2014)) was applied across China to estimate triclosan removal in wastewater treatment in this study. The annual emission of triclosan for counties was estimated by combining the urban and rural usage, wastewater treatment connection rates and the ingredient removal efficiency in wastewater treatment (Eq. (2) in SI) for the five economic scenarios.

2.5. Generation of concentrations

Triclosan concentrations in surface waters across China were modelled using the Sino Evaluative Simplebox-MAMI Model (SESAMe) v3.3 (Zhu et al., 2016; Zhu et al., 2015), 50 km grid multimedia chemical fate model taking into account of chemical ionization, for 2000–2030 under the five economic scenarios, combined with different surface water discharge flow scenarios. The model configuration with input parameters and the chemical properties are detailed in the SI. Table S3 shows the amount of freshwater applied on agricultural irrigation for each province or municipality. The model has been validated on estimating triclosan concentrations in China in a 2012-year scenario and performed well in a previous study (Zhu et al., 2016). The thirty years' emission by counties estimated in the last step was interpolated by population to fit the 50 km grid for the model by ArcGIS 10.4. Future discharge flows predicted by Global Water Availability Assessment model (GWAVA) (Meigh et al., 1999) were acquired under two IPCC marker scenarios (A2 and B1), and the two marker scenarios were modelled by a general circulation model, ECHAM5 model (WATCH, 2011). In this study, the A2 discharge flow was assigned to the two 'A' economic scenarios and the B1 discharge flow was assigned to the two 'B' and the OECD economic scenarios as environmental variables in SESAMe v3.3 for 2011–2030. The estimated historical average discharge flow used in a previous study (Zhu et al., 2016) was assigned for 2000–2010. To reflect the influence of the discharge dilution on the water concentration, the historical average discharge flow was again applied all through 2000–2030 under the five economic scenarios for comparison. This is called the "constant discharge flow" below to differentiate with the A2 and B1 discharge flows. The change in other environmental variables through 2000–2030 was not considered, as their influence to the surface water concentration of triclosan was limited (Zhu et al., 2014).

2.6. Ranking the importance of drivers to chemical concentration changes in surface water

A variance-based sensitivity analysis, Sobol's global sensitivity analysis (Saltelli et al., 2008; Zhu et al., 2014) was applied to capture the influence of the full range of spatial and temporal variation of the main drivers to the surface water concentration of triclosan for the whole country. The total-order index was calculated to rank the importance of drivers, which is the total sensitivity of the individual driver contributed by the single driver and its interaction with other drivers (joint effects). A larger index indicates greater importance (see SI).

The six main drivers were: population; urbanization rates; wastewater treatment connection rates for rural and urban areas, respectively; per capita GDP and surface water discharge flows (dilution). For the other environmental parameters, the surface water concentrations are not very sensitive to them and their temporal change is not available yet (Zhu et al., 2014). The sensitivity analysis was conducted twice, for the periods 2000–2010 and 2010–2030 respectively, under only the OECD economic scenario, as the changing rate of these drivers may be different in the two periods. For example, urban wastewater treatment connection rates will have grown faster in the first ten years than in the later twenty years, while development of rural wastewater treatment facilities have been faster after 2010 than before (Fig. 2A) and is expected to grow in such quicker speed due to a currently stable investment and focus by the government (MHURD, 2013). Values of parameters for the grid cells in the SESAMe v3.3 model across mainland China for the first ten years and the later twenty years made two original datasets for the six drivers. The Latin Hypercube sampling method (Zhang and Pinder, 2003) was used to take 10,000 values from the original datasets for each driver to compose random datasets for the sensitivity analysis. Such sampling method could make sure that the random values cover the full range of parameter values more evenly. For more details on this sensitivity analysis method see the section S5 in SI.

3. Results and discussion

3.1. Population and urbanization trends

From population projections, China's total population increased by ca. 5% in ten years from 2000 (1.26 billion) to 2010 and is predicted to increase by ca. 6% in twenty years from 2010 to 2030 (1.41 billion). What is more dramatic is the change in urbanization rates. We estimate this to be as follows nationally: 2000, 31.6%; 2010, 48.5%; 2020, 48.2%; 2030, 74.7% (SI Table S4) - a huge shift over the study decades. Compared to the historical data (2000–2015) in yearbooks (CNSTATS 2000–2015), our estimation is slightly lower, but the trends are very clear (SI Fig. S4). Differences between estimates are probably influenced by the assumptions for the threshold-value method used for urban population density, i.e. one identical value for the whole country and the choice of threshold values. The urbanization rate by 2030 predicted by this study is close to the projection by the World Bank and the Development Research Center of the State Council, China (70%) (Guthrie, 2007). From this estimate, the annual growth rate of the urban population is ~1.9% from 2015 to 2030.

3.2. Wastewater treatment connection rates

China has made huge investments in urban wastewater treatment infrastructure over recent years. After 2000, the national average urban wastewater treatment connection rate grew fastest during 2003–2010, increasing about 2.8 times in just 7 years; and projections in this study suggest slower but substantial increases during 2010–2030 (Fig. 2 and SI Fig. S1 and S5). At the beginning of the study period, there was large regional variation for urban areas, but this is reducing as modernisation continues across China (Fig. 2B and SI Fig. S5). For example, by 2010 the provincial average urban wastewater treatment connection rate was < 50% in Heilongjiang and Qinghai; but in contrast, it was > 85% in Chongqing, Tianjin, Hebei, Jiangsu, Anhui and Shandong. However, by 2015, the lowest provincial urban wastewater treatment connection rate had already reached about 80%. In the outline of "13th Five-year Plan", the urban wastewater treatment connection rates in cities and county towns (Xian Cheng) are expected to reach 95% and 85% respectively by 2020 (CPGC, 2016). From projections by this study, the national urban wastewater treatment connection rate will be

96 ± 17% (mean ± STD (standard deviation)) by 2020 (SI Table S5). This is an average of cities and county towns without weighting by population. Almost all provinces will be close to or beyond the national target of 95% by 2020 except for Hainan province (90%) predicted by this study.

We could not find any records of the extent of rural wastewater treatment before 2008, so assumed a low and relatively stable wastewater treatment connection rate (1.9–2.7%) for 2000–2007 for rural areas. After 2010, the national average rural wastewater treatment grows faster and is projected to grow about 24 times during 2010–2030 (Fig. 2A). Provincial level variation is low and relatively stable before 2010 but estimated to increase thereafter (SI Table S5). For example, during 2015–2030 rural wastewater treatment facilities are projected to develop much faster in Sichuan, Chongqing, Shanxi, Qinghai, Shaanxi, Ningxia and some coastal provinces than other regions, especially Inner Mongolia and Jiangxi, than in other provinces (SI Fig. S6). The Chinese government plans to promote rural wastewater treatment infrastructures gradually (CPGC, 2016) but there has not been a specific numerical target such as that for urban areas. However half of the Chinese population are living in rural areas, and considering that in rural villages wastewater produced is normally 60–80% of the water usage and closely linked to the water usage within the same village (Godavitarne et al., 2011), it is crucial to develop infrastructures for wastewater treatment in rural areas.

3.3. Projected usage and emissions under five economic scenarios

The increase in estimated usage follows that of per capita GDP for the five economic scenarios during 2010–2030 (Fig. 3A and SI Fig. S3). Usage is predicted to grow relatively moderately under the OECD, B1 and B2 economic scenarios, more sharply after 2020 in the A1 economic scenario and relatively slowly in the A2 economic scenario. Triclosan usage in 2000 and 2010 is estimated to be ca. 86 and 160 t, respectively. Under the OECD economic scenario, its usage is estimated to increase about twofold over the twenty years, to reach ~340 t in 2030. Under the A1 economic scenario, usage would increase ~3.7 times during 2010–2030, to reach ~590 t p.a. The relative projected emissions for the five economic scenarios is similar to that for the usage, i.e. emissions are the highest and the lowest respectively under the A1 and A2 economic scenarios, and moderate in the other three economic scenarios. However, the change of emissions with time under the five economic scenarios is more complex than the usage (Fig. 3B) due to being driven by the combination of several factors and variation in data sources for different points in time. The detailed annual usage is in SI Table S6.

The percentage of total triclosan mass removed by all wastewater treatment infrastructures in China is projected to increase from 11% to 85% during 2000–2030 (SI Fig. S7) as a result of urbanization and the development of wastewater treatment infrastructures all over China. National total emissions are projected to be highest in 2007 (100 t) and decrease from ca. 76 to 43–57 t in 2030 under four economic scenarios except A1 (SI Table S7). Under A1 economic scenario, triclosan emissions remain relatively high and reach the peak in 2025 (110 t), with the projected increased usage after 2020 being somewhat offset by increased removal with urbanization and infrastructure developments. There is an apparent sharper change in emissions in 2025 than in other years in the study period (Fig. 3B). This is an artefact of the approach, because we had to use different population data sources for 2020 and 2030, as explained above.

SI Figs. S8–S9 show spatial distributions of triclosan usage and emissions at the county scale for every five years from 2000 to 2030 under the OECD economic scenario. The distribution of usage is determined by population distribution as stated above. Therefore, most usage over the whole 2000–2030 period is into eastern or southeast China below the Heihe-Tenchong Line, a geo-demographic demarcation line that divides the area of China roughly into two equal parts with about 94% of the Chinese population in the west (SI Fig. S8 for 2000). This covers almost half of China's land mass area, but is occupied by ~94% of China's population (surveyed for 2002) (Guthrie, 2007). In the west, the usage in some counties in Xinjiang is estimated to be high. Lowest usage is estimated to be in Tibet. Generally, the spatial distribution of emissions is very similar to that of usage for 2000–2010. However, after 2015 emissions are projected to decline steadily in Beijing, southern Hebei, Chongqing, eastern Sichuan, eastern coastal cities or provinces such as Shanghai, Zhejiang and Guangdong. Until 2030, emissions in mid-Heilongjiang, south Jilin, east Liaoning, most areas of Hebei, Beijing, Tianjin, Shanxi, Shandong and most areas in south China are predicted to be rather low and below 0.008 t per annum in each county. Most counties in Inner Mongolia and some counties in Xinjiang, eastern Henan and northern Anhui are still estimated to have high emissions. This is because of low urbanization rates and low rural wastewater treatment connection rates projected for these areas in 2030.

3.4. Projected concentrations

Triclosan concentrations in surface water are predicted by the SESAMe v3.3 model with ~50 km spatial resolution. Fig. 4 and SI Fig. S10 shows temporal change of the national median concentrations for the 50 km grid cells as a result of the combination of different economic and discharge scenarios. During 2011–2030, the median concentration is estimated to be highest under economic scenario A1 and lowest under A2, and moderate in B1, B2 and OECD economic scenarios. The pattern is similar to that of usage and emissions (Fig. 3). When applying different discharge flow scenarios for 2011–2030, the A2 and B1 discharge flows possibly would dilute the chemical in surface water more during 2011–2025 but less during 2026–2030 compared to the constant discharge flow. SI Fig. S11-S13 show the histogram of predicted triclosan concentrations in surface water during 2000–2030 under different scenarios. Fig. S12–S13 indicates that a wider range of triclosan concentrations across China would be estimated by using the constant discharge flow rather than using A2 and B1 discharge flows under all

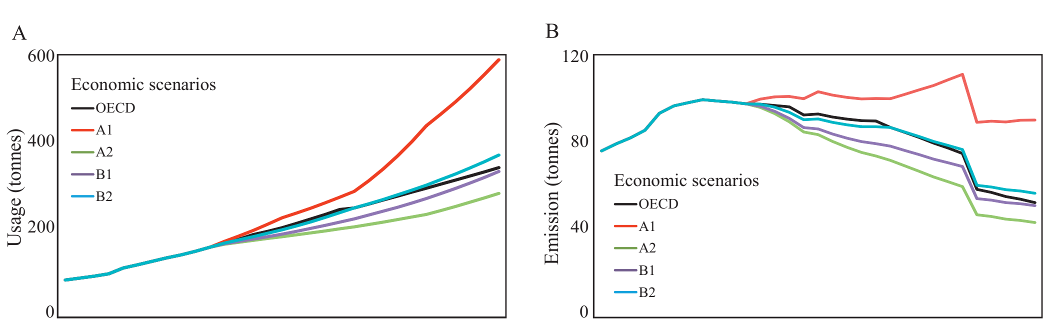

Fig. 3. Projected annual total usage (A) and emissions (B) of triclosan in mainland China from 2000 to 2030 under five economic scenarios.

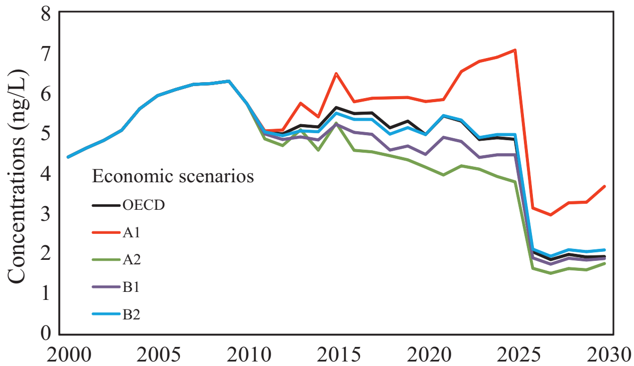

Fig. 4. Predicted national median concentrations from 2000 to 2030 under five economic scenarios; the estimated constant discharge flow for 2000–2010 is used for 2000–2010, while for 2011–2030 the A2 discharge flow is used in A1 and A2 economic scenarios and B1 discharge flow is used in B1, B2 and OECD economic scenarios. Note: the estimated median concentration has an apparent sharp decline in 2025, as noted earlier for the emission projects.

five economic scenarios.

When applying A2 and B1 discharge flows for 2011–2030, the highest median concentration of triclosan is estimated to be in 2009 (ca. 6.3 ng/L) under OECD, A2, B1 and B2 economic scenarios, and in 2025 (ca. 7.0 ng/L) under A1 economic scenario (Fig. 4). The median concentration generally declines after 2009 in all economic scenarios except A1. When applying the constant discharge flow for 2011–2030, the highest median concentration of triclosan is estimated to be in 2016 under OECD (~6.7 ng/L) and B2 (~6.6 ng/L) economic scenarios, in 2025 under the A1 (~8.2 ng/L) economic scenario, and in 2009 under the A2 and B1 (~6.3 ng/L) economic scenarios (Fig. S10). The estimated median concentration has a sharp decline in 2025, which is the same as the emission due to the same reason discussed above. It has declined to ca. 3–3.5 ng/L for A1 economic scenario and ca. 1.5–2 ng/L for all other economic scenarios.

Fig. 5 shows the temporal evolution pattern of triclosan concentration frequency in water with different discharge scenarios under the OECD economic scenario. Generally, regions with relatively high concentrations increase after 2000 and then decrease in 2020 and 2030. Regional concentrations are more scattered when applying constant discharge flow (Fig. 5B) than applying B1 discharge flow (Fig. 5A), as more regions having concentrations < 1 ng/L or > 100 ng/L are estimated when applying constant discharge flow in the model prediction. 100 ng/L is the current UK maximum guideline value for total triclosan in surface water (UKTAG, 2015). Proportion of areas with estimated concentrations > 100 ng/L is about 10% in the four years using the constant discharge flow (Fig. 5B), but decreases to only ~2% when using the B1 discharge flow in 2020 and 2030 (Fig. 5A).

SI Figs. S14–S15 show the spatial distribution of concentrations estimated under the OECD economic scenario using the B1 discharge flow and the constant discharge flow. When applying the constant discharge flow, estimated concentrations in most regions in Shanxi, North China Plain (NCP), mid-Inner Mongolia and part of the three provinces in the northeast are relatively high all through 2000–2030, due to the low surface water dilution. Estimated concentrations in South China are lower than above regions and decline steadily from 2015 to 2030 (SI Fig. S15). Such spatial distribution patterns should also be obtained under other economic scenarios, as the constant discharge flow is applied identically. When applying the B1 discharge flow for 2015–2030, estimated concentrations in above high concentration areas are still higher than the other areas, but decline significantly after 2015. Compared to the extent of national median concentration change during the study period, i.e. about threefold (Fig. 4), the extent of the regional concentration change could reach over 5000 times as much during the period in Zhada, Tibet. The average concentration of triclosan in surface water for15% of counties across China has changed over 50 times as much over the thirty years.

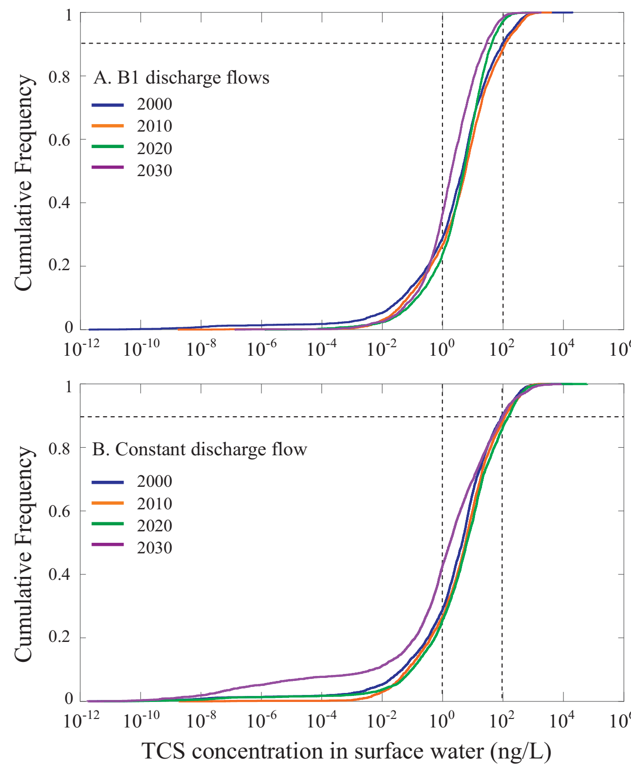

Fig. 5. The cumulative frequency of triclosan concentrations in surface water under the OECD economic scenario for 2000, 2010, 2020 and 2030. The constant discharge flow is used for 2000 and 2010 in both figures. A. B1 discharge scenario used for 2020 and 2030; B. the constant discharge flow used for 2020 and 2030.

The results on ingredient usage, emissions and concentrations are illustrated and discussed for triclosan. Different temporal patterns for different HPCP ingredients may occur, but the methodology for estimations is adapted.

3.5. Contrasting the spatial distribution of usage, emissions and concentrations

Using the B1 discharge flow here for discussion, the spatial distributions of projected usage, emissions and concentrations show geographic contrasts, because of the influence of regional urbanization rates, wastewater treatment connection rates and surface water dilution. For example, the estimated highest usage is in urban Dongguan, Guangdong province from 2000 to 2025 and in Tailai, Heilongjiang from 2026 to 2030. However, the estimated highest emission is in counties in Guandong (Dongguan or Puning) during 2000–2010, in Yunnan (Xuanwei) during 2011–2025 and in Henan in 2030. Meanwhile, the estimated highest concentrations are in Shanxi (Yingze) during 2000–2010, but in Hebei (Shanhaiguan) during 2011–2030.

The estimated top five counties for the highest usage change from mostly in Guangdong to Beijing (Chaoyang and Haidian districts) and Shanghai (Pudong New Area), and then to Chongqing (Yubei, Yongchuan and Jiulongpo districts) over the 30 years. In contrast, the top five counties that have the highest emission across China change from mostly in Guangdong until 2010 to counties mainly in the southwest, such as in Guangxi (Guiping and Bobai), Guizhou (Weining Yi, Hui and Miao autonomous county) and Yunnan (Xuanwei and Zhenxiong) until 2025, and change to counties all in Henan in 2026–2030. The estimated top five counties that have the highest concentration are mostly in Shanxi from 2000 to 2010 and change to mostly in Hebei from 2015 to 2030. SI Table S8 shows the top five

counties with the estimated highest usage, emission and concentration for 2000, 2010, 2020 and 2030 for comparison.

SI Fig. S16 illustrates the temporal change of average wastewater treatment connection rates for 11 provinces/cities over 2000–2030. As it is urbanization rates and population weighted (See SI and SI Eq. 3), it provides direct reasons for the contrast between usage and emissions as illustrated above. Beijing and Shanghai have relatively high estimated wastewater treatment connection rates throughout the 30 years and therefore do not have the highest emission in terms of the highest usage during the same period. Wastewater treatment connection rates in Yunnan, Guangxi, Guizhou and Heilongjiang are estimated to be much lower and grow slowly compared to other provinces during 2000–2025, but increase sharply to different extent after 2025. Therefore emissions are estimated to be the highest in some counties in these provinces during 2010–2025. The average wastewater treatment connection rates in Henan are estimated to decline after 2025 and be lower than that in Heilongjiang, so emissions in counties there turn to the highest in 2030. The contrast between the spatial distribution of emissions and concentrations is a result of the effect from surface water discharge flow.

3.6. Ranking of different drivers

The coefficient of variation (CV) describes the amount of variability relative to the mean (STD/mean). In Fig. 6A, CV (bars in the figure) considers both the spatial variation (5469 grid cells across mainland China) and the temporal (annually during 2000–2010 and 2010–2030) change of all projected drivers during the two periods. The percentage values (%) above the bars reflect the temporal variation only at national average level. The CV of discharge flows (highest bars marked with Q) and population is mainly contributed by the spatial variation due to the extremely low % values above the bars, and that of per capita GDP is contributed only by temporal change. Discharge flow has the highest variation for the two periods. For the other five drivers, the rank of the variation is in order of population, urbanization rates, urban wastewater treatment connection rates, rural wastewater treatment connection rates and per capita GDP for 2000–2010; and population, rural wastewater treatment connection rates, urbanization rates, urban wastewater treatment connection rates and per capita GDP for 2010–2030. Comparing the variation in the two periods, only projected urban wastewater treatment connection rates and discharge flows have higher variation in 2000–2010 than in 2010–2030. The other four projected drivers all have higher variation in 2010–2030 especially for rural wastewater treatment connection rates. It reveals that the development of urban WWTPs during 2010–2030 is estimated to be moderated and approach complete in a national wide compared to the previous ten years; whilst the other four drivers are estimated to have more dramatic development (e.g. rural wastewater treatment facilities and per capita GDP) or spatial variation (e.g. population distribution and urbanization rates) in 2010–2030 than previous years.

The total-order index of the global sensitivity analysis in this study (Fig. 6B) reflects the relative importance of changes of the six drivers to triclosan concentrations in surface water over the study periods across China taking account of interactions between drivers. The regional population and discharge flow are projected to be the most important drivers to changes in triclosan concentrations for both periods, as the regional population determines the spatial allocation of the usage and discharge flow affects the dilution directly. For the other four drivers in Fig. 6B, during 2000–2010, the development of urban wastewater infrastructures and the urbanization progress are projected to be more important to triclosan concentrations across China, followed by rural wastewater treatment infrastructure development and per capita GDP growth in sequence. Contrarily, during 2010–2030, the development of rural wastewater treatment infrastructure probably becomes the most important among the four drivers to triclosan concentrations for China, followed by per capita GDP growth, urban treatment infrastructure development and urbanization progress in sequence. The difference of the estimated relative importance is more significant during 2010–2030 than 2000–2010 for the four socioeconomic drivers possibly due to the more significantly different variation of the drivers during 2010–2030 (Fig. 6A).

The rank of the importance of the drivers to triclosan concentrations does not exactly follow that of variations of the drivers, as (1) it is a non-linear model and (2) changes of multiple drivers will influence triclosan concentrations jointly (interactions of drivers as mentioned above). This implies that it is not reasonable to compare the influence of several changing drivers to surface water concentration of a chemical by simply comparing the extent of the variation/change of drivers. However, if a driver develops more dramatically during a period than any other time, e.g., rural wastewater treatment connection rates during 2010–2030, its relative importance might increase during this period. The global sensitivity analysis was conducted based on triclosan. However, as these main drivers are mostly relevant to sources of the ingredient rather than chemical properties, the conclusion can be generalized to a range of down-the-drain HPCP ingredients that have similar sources with triclosan.

3.7. Uncertainties

Uncertainties of the methodology in this study exist and are common for similar source HPCP ingredients. The uncertainties of using the threshold-value method to identify the urban population have been discussed above. Emissions might be slightly overestimated as a result of following assumptions: (1) The allocation method is applied without differentiating the purchasing power in urban and rural areas. In reality, per capita usage of HPCPs in urban areas might be higher than that in rural areas due to the higher GDP levels in urban areas. Therefore more HPCP ingredients should have been removed as a result of higher wastewater treatment connection rates in urban areas than rural areas.

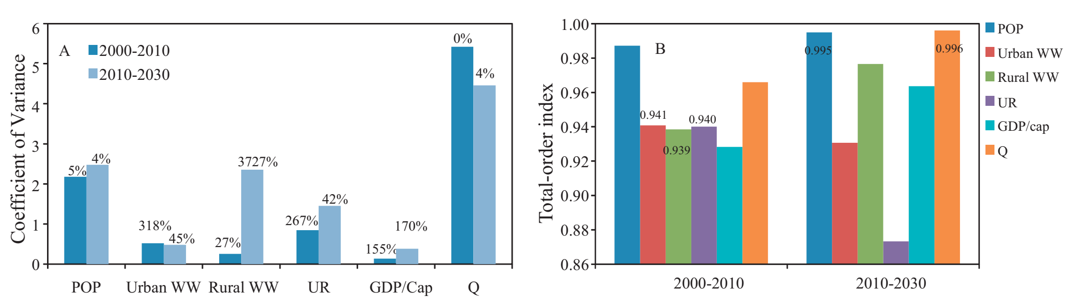

Fig. 6. A. coefficient of variation of values for 50 km grid of the six drivers across China during 2000–2010 and 2010–2030; the values above the bars are percentages of national average driver changes from the start to the end year for each period. B. total-order index from the sobol's global sensitivity analysis for respective 2000–2010 and 2010–2030. POP, population; Urban/Rural WW, urban/rural wastewater treatment connection rates; UR, urbanization rate; GDP/Cap, per capita GDP (Gross Domestic Product); Q, surface water discharge flow. The projection is under OECD (Organisation for Economic Co-operation) economic scenario for 2000–2030 and constant discharge flow for 2000–2010, B1 discharge flow for 2011–2030. To explicit about their relative importance, the exact values of total-order index are added in fig. B for several parameters for which the values are very close to each other.

(2) No bans or elimination are assumed in this study till 2030 in China. However, formula change by replacement or reduction of ingredient use is possible if there are restrictions by government or if it is phased out by industry. For triclosan, there is an indication that industry are beginning to phase this ingredient out now. (3) a high level ingredient inclusion level is assumed to be identical in all HPCP categories. In some categories it is actually lower than this. (4) An identical removal efficiency for typical secondary activated sludge plants is applied across China for all wastewater treatment infrastructures. It might overestimate the removal in some rural areas where only primary treatment facilities are used.

Slight underestimation of emissions might be caused by the following assumptions: (1) Only sales data of HPCP ingredients that are released down to drain with domestic wastewater is accounted for in this study. Many HPCP ingredients are at the same time contained in solid materials, such as triclosan used in plastic and textiles, might be transported to aquatic system with runoff after the products are disposed in land. However, it might be very slow release. (2) Sludge application in soils is not considered in this study. (3) Possible release from processes of manufacture of HPCPs is not considered. It might exist in some regions but be rather low compared to those accounted in this study.

Other uncertainties may also exist. For example, (1) spatial allocation by population might cause skewed spatial distribution of HPCP usage, as the usage might be more relevant to regional GDP levels. (2) As explained above, the relatively sharp change of emissions and national median concentrations in 2025 might be caused by different population data sources. It also reflects uncertainties of future population distribution. The projections from the different sources may be based on distinct assumptions, but both are possible. (3) The projection of population and migration from sources cited in this study probably has not considered the end of one-child policy and permission of a second child for a couple effective in 2016. Such national policy or other economic policies in regional or national scale may all affect future population count and distribution. (4) Per capita GDP might be more important than the prediction in this study considering its spatial variation in reality.

4. Conclusions and implications

For the first time, a methodology is presented for assessing possible future water concentration of contaminants for China under different economic scenarios and the key drivers, which will be useful to policy-makers and stakeholders for mitigation of domestic effluents affecting water quality. Here triclosan is only chosen as an example HPCP ingredient, and the predicted concentrations in this study would be advisable to embark on monitoring work over following a few years to confirm the reliability of this study. All other chemicals which mainly derive from anthropogenic sources that are relevant to economic levels are suitable for this method. HPCPs as trace organics can be marker chemicals for similar anthropogenic-source chemicals, of which the usage is linked to GDP. This framework can provide a preliminary outlook of surface water contamination levels of these chemicals and therefore provide information for chemical management in China.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2018.03.013.

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